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### (54) **Crystalline propylene block-copolymer composition for use in automobile parts**

Kristalline Propylen-Blockcopolymerzusammensetzungen für Kraftfahrzeugteile

Compositions de propylène copolymères séquencés utilisées dans des pièces d'automobile

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(56) References cited:

<b>EP-A- 0 463 963</b>	<b>EP-A- 0 509 662</b>
<b>EP-A- 0 566 141</b>	<b>EP-A- 0 697 435</b>
<b>EP-A- 0 794 225</b>	<b>US-A- 5 045 589</b>
<b>US-A- 5 543 454</b>	

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**Description****FIELD OF THE INVENTION**

5 [0001] The present invention relates to a crystalline propylene block-copolymer composition for use in automobile parts, which comprises a crystalline propylene block-copolymer, elastomers and talc. More specifically, it relates to a crystalline propylene block-copolymer composition for use in automobile parts, which is superior in the flowability upon injection molding.

**BACKGROUND OF THE INVENTION**

[0002] Polypropylene has found wide applications in various fields including household commodities, kitchen utensils, packaging films, automobile parts, machine parts and electric appliances, in which additives are incorporated in accordance with each specific required performance. For example, polypropylene compositions containing such additives as an elastomer and talc are employed in application fields in which mechanical strength is required, such as automobile parts.

[0003] There has, however, been expected an improvement in the flowability upon injection molding for the polypropylene compositions of prior art exhibiting improved material properties by the addition of an elastomeric component and a talc component (see eg EP-A-O 697 435).

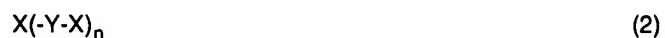
**SUMMARY OF THE INVENTION**

[0004] An object of the present invention is to provide a crystalline propylene block-copolymer composition for use in automobile parts, which is superior in the flowability upon molding and also in the balance between the physical properties, such as flexural modulus, impact resistance, hardness and brittle temperature.

[0005] The crystalline propylene block-copolymer composition for use in automobile parts according to the present invention, comprises

(A) 41 to 71 % by weight of a crystalline propylene block-copolymer composed of a propylene homopolymer portion (Fp) and an ethylene/propylene random copolymer portion (Fc) and having an ethylene content of 1 to 10 % by weight and a melt flow rate (MFR, determined at 230 °C under a load of 2160 g) of 20 to 100 g/10 min.,

(B) 12 to 22 % by weight of a first hydrogenated product of a block-copolymer composed of blocks represented by the following formula (1) or (2), namely,



in which X stands for a polymer block of a vinyl-mono-substituted aromatic hydrocarbon, Y stands for an elastomeric polymer block of a conjugated diene and n is an integer of 1 to 5, wherein the content of X amounts to 5 to 22 % by weight and the melt flow rate (MFR) for the entire block-copolymer is 0.5 to 70 g/10 min. and wherein the hydrogenation degree is at least 90 mole %,

(C) 2 to 12 % by weight of a second hydrogenated product of a block-copolymer composed of blocks represented by the following formula (1) or (2), namely,



in which X stands for a polymer block of a vinyl-mono-substituted aromatic hydrocarbon, Y stands for an elastomeric polymer block of a conjugated diene and n is an integer of 1 to 5, wherein the content of X amounts to 30 to 80 % by weight and the melt flow rate (MFR) for the entire block-copolymer is 0.1 to 20 g/10 min. and wherein the hydrogenation degree is at least 90 mole %, and

(D) 15 to 25 % by weight of talc of a grade having an average particle size, determined by a laser beam diffraction method, of 1 to 5  $\mu\text{m}$ ,

wherein, in the crystalline propylene block-copolymer (A), the said propylene homopolymer portion (Fp) contains 20 to 50 % by weight of a low molecular weight propylene homopolymer (Fh) having a melt flow rate (MFR, determined at 230 °C under a load of 2160 g) of 400 to 1,000 g/10 min. and is constituted of a propylene homopolymer having an average melt flow rate (MFR, determined at 230 °C under a load of 2160 g), for the entire propylene homopolymer portion (Fp), of 100 to 350 g/10 min. and an isotactic pentad fraction (mmmm), determined by  $^{13}\text{C}$ -NMR, of at least 98 % and the said ethylene/propylene random copolymer portion (Fc) is constituted of an ethylene/propylene random copolymer having an intrinsic viscosity  $[\eta]$ , determined at 135 °C in decalin (decahydronaphthalene), of 7 dl/g or higher.

#### DETAILED DESCRIPTION OF THE INVENTION

**[0006]** The crystalline propylene block-copolymer (A) to be incorporated according to the present invention is constituted of a block-copolymer composed of a propylene homopolymer portion (represented hereinafter sometimes by Fp) and an ethylene/propylene random copolymer portion (represented hereinafter sometimes as Fc). The ethylene content in the entire block-copolymer (A) amounts to 1 to 10 % by weight, preferably 3 to 8 % by weight, and the melt flow rate (MFR, determined in accordance with ASTM D 1238 at 230 °C under a load of 2160g; the same applies in the following) of the entire block-copolymer (A) is 20 to 100 g/10 min., preferably 30 to 90 g/10 min.

**[0007]** The propylene homopolymer portion (Fp) composing the crystalline propylene block-copolymer (A) is a homopolymer of propylene having an isotactic pentad fraction (mmmm) of at least 98 %, preferably at least 98.5 %.

**[0008]** The above-mentioned isotactic pentad fraction corresponds to the proportion of the isotactic chains as the pentad unit in the molecular chains of the crystalline propylene copolymer (A), which is determined using  $^{13}\text{C}$ -NMR and which is the proportion of the number of propylene monomeric units present in each center of the sequences of 5 monomeric units bound each other successively by meso-coupling. It is determined practically as the proportion of the mmmm peaks in the entire absorption peaks within the methyl-carbon region of  $^{13}\text{C}$ -NMR spectrum.

**[0009]** In general, a polymer material does not consist of an aggregate of molecules of same structure and same molecular weight but is a mixture of polymer molecules of different molecular weights, which exhibits certain material properties as a whole. The crystalline propylene block-copolymer (A) to be incorporated in the resin composition according to the present invention is also an aggregate of polymer molecules of different molecular weights and exhibits, with respect to the integrated polymer material, an ethylene content and an MFR as specified above, wherein further restrictions are incorporated according to the present invention in the propylene homopolymer portion (Fp) composing the block-copolymer (A) as to the molecular weight and the content thereof. Thus, the propylene homopolymer portion (Fp) has an MFR (average MFR for the entire homopolymer portion) of 100 to 350 g/10 min., preferably 150 to 300 g/10 min., and contains 20 to 50 % by weight, preferably 25 to 40 % by weight, of a low molecular weight propylene homopolymer (Fh) having an MFR of 400 to 1,000 g/10 min., preferably 400 to 800 g/10 min., based on the weight of the entire propylene homopolymer portion (Fp). MFR serves as an indicator for the molecular weight, wherein the greater the value of MFR, the smaller would be the molecular weight.

**[0010]** The ethylene/propylene random copolymer portion (Fc) composing the crystalline propylene block-copolymer (A) has an intrinsic viscosity  $[\eta]$ , determined at 135 °C in decalin, of at least 7 dl/g, preferably 8 to 15 dl/g.

**[0011]** The ethylene content in the ethylene/propylene random copolymer portion (Fc) may desirably amount to 15 to 50 % by weight, preferably 20 to 30 % by weight.

**[0012]** The content of the propylene homopolymer portion (Fp) in the crystalline propylene block-copolymer (A) may favorably amount to 85 to 99 % by weight, preferably 90 to 97 % by weight and the content of the ethylene/propylene random copolymer portion (Fc) in the crystalline propylene block-copolymer (A) may favorably amount to 1 to 15 % by weight, preferably 3 to 10 % by weight.

**[0013]** It is permissible that only one single product of crystalline propylene block-copolymer (A) is employed or two or more products thereof are used in a combination.

**[0014]** The crystalline propylene block-copolymer (A) can be produced in various ways, for example, using a known stereospecific catalyst. In practice, it can be produced by effecting a block-copolymerization in the presence of a catalyst formed from a solid titanium catalyst component, an organometallic compound catalyst component and, if necessary, an electron donor. Alternatively, it can be produced by effecting a block-copolymerization in the presence of a lithium catalyst or a Ziegler catalyst in an inert solvent. Details of such production procedures are disclosed in, for example, Japanese Patent Publication Sho 40-23798 B.

**[0015]** The first hydrogenated product of block-copolymer (B) to be incorporated in the composition according to the present invention is a hydrogenated product of a block-copolymer composed of blocks represented by the formula (1) or (2) given previously, wherein the content of the blocks of X amounts to 5 to 22 % by weight, preferably 10 to 20 %

by weight, and the melt flow rate (MFR, determined at 230 °C under a load of 2160 g) for the entire block-copolymer is 0.5 to 70 g/10 min., preferably 1 to 30 g/10 min. The degree of hydrogenation is at least 90 mole %, preferably at least 95 mole %.

[0016] As the vinyl-mono-substituted aromatic hydrocarbon constituting the polymer block represented by X in the formula (1) or (2), there may be enumerated styrene and derivatives thereof, for example, styrene,  $\alpha$ -methylstyrene, p-methylstyrene, chlorostyrene, lower alkyl-substituted styrenes and vinyl-naphthalene. They may be incorporated either alone or in a combination of two or more of them.

[0017] As the conjugated diene constituting the elastomeric polymer block represented by Y in the formula (1) or (2), there may be enumerated, for example, butadiene, isoprene and chloroprene. They may be incorporated either alone or in a combination of two or more of them. n is an integer of 1 to 5, preferably 1 or 2.

[0018] Concrete examples of the block-copolymer for the first hydrogenation product (B) include those block-copolymers based on styrene which satisfy the condition for the material properties as given previously, such as styrene/ethylene-butene/styrene triblock-copolymer (SEBS), styrene/ethylene-propylene/styrene triblock-copolymer (SEPS) and styrene/ethylene-propylene diblock-copolymer (SEP). The first hydrogenation product of block-copolymer (B) may be incorporated either alone or in a combination of two or more of them.

[0019] The block-copolymer which is to be hydrogenated afterwards can be produced by, for example, effecting a block-copolymerization in an inert solvent in the presence of a lithium catalyst or a Ziegler catalyst. Details of such production procedures are disclosed in, for example, Japanese Patent Publication Sho 40-23798 B.

[0020] The hydrogenation treatment can be carried out in an inert solvent in the presence of a known hydrogenation catalyst. Details of such production procedures are disclosed in, for example, Japanese Patent Publications Sho 42-8704 B, Sho-43-6636 B and Sho-46-20814 B.

[0021] The second hydrogenation product of block-copolymer (C) is a hydrogenated product of a block-copolymer composed of blocks represented by the formula (1) or (2) given previously, wherein the content of X amounts to 30 to 80 % by weight, preferably 40 to 70 % by weight, and the melt flow rate (MFR, determined at 230 °C, under a load of 2160 g) for the entire block-copolymer is 0.1 to 20 g/10 min., preferably 0.3 to 15 g/10 min. The hydrogenation degree is at least 90 mole %, preferably at least 95 mole %.

[0022] As the vinyl-mono-substituted aromatic hydrocarbon constituting the polymer block X and as the conjugated diene constituting the elastomeric polymer block Y in the formula (1) or (2), those which are exemplified as for the first hydrogenated product of block-copolymer (B) are exemplified. n is an integer of 1 to 5, preferably 1 or 2.

[0023] Concrete examples of the second hydrogenation product of block-copolymer (C) include those block-copolymers based on styrene which satisfy the condition for the material properties given previously, such as styrene/ethylene-butene/styrene triblock-copolymer (SEBS), styrene/ethylene-propylene/styrene triblock-copolymer (SEPS) and styrene/ethylene-propylene diblock-copolymer (SEP).

[0024] The second hydrogenation product of block-copolymer (C) may be incorporated either alone or in a combination of two or more of them.

[0025] The second hydrogenation product of block-copolymer (C) can be produced in the same manner as in the first hydrogenation product of block-copolymer (B).

[0026] The talc (D) to be incorporated in the composition according to the present invention is of a grade in which the average particle size determined by a laser beam diffraction method is in the range from 1 to 5  $\mu$ m, preferably from 1 to 3  $\mu$ m. Any grade of talc having an average particle size falling under the above range can be employed without any restriction.

[0027] The proportion of each component of the composition according to the present invention amounts to 41 to 71 % by weight, preferably 52 to 66 % by weight, for the crystalline propylene block-copolymer, 12 to 22 % by weight, preferably 13 to 19 % by weight, for the first hydrogenation product of block-copolymer (B), 2 to 12 % by weight, preferably 2 to 6 % by weight, for the second hydrogenation product of block-copolymer (C), and 15 to 25 % by weight, preferably 18 to 22 % by weight, for talc (D).

[0028] It is permissible, if necessary, to incorporate in the composition according to the present invention other additives, such as heat stabilizer, antistatic agent, UV-absorber, light stabilizer, aging inhibitor, antioxidant, softening agent, dispersing agent, filler, pigment and lubricant, in addition to the components (A) to (D), within the range not obstructing the purpose of the present invention.

[0029] The composition according to the present invention can be obtained by blending or melt-mixing the constituent components (A) to (D) and the additives to be incorporated on requirement on a mixing apparatus, such as Bumbury's mixer, a single-screw extruder, a twin-screw extruder or a high speed twin-screw extruder.

[0030] The composition according to the present invention obtained as above is superior in the flowability upon molding thereof and also in the balance between the material properties, such as flexural modulus, impact resistance, hardness and brittle temperature. For this reason, the composition according to the present invention can favorably be utilized as a starting material of the resin for automobile parts and, in particular, as the raw resin material for injection molding, wherein it exhibits quite better flowability upon the injection molding, whereby a molded article can be produced

easily.

**[0031]** Concrete examples of such injection-molded automobile parts include automobile exterior parts, such as bumper, side molding, garnish, window molding and wheel house cover; and interior parts, such as instrument panel, glove box, console box, door trim, pillar trim and steering column cover.

**[0032]** Due to the specifically proportioned contents of the specific crystalline propylene block-copolymer, the specific hydrogenated products of block-copolymers and the specific grade of talc, the crystalline propylene block-copolymer composition for use in automobile parts according to the present invention is superior in the flowability upon molding thereof and in the balance between the physical properties, such as flexural modulus, impact resistance, hardness and brittle temperature.

#### MODE FOR EMBODYING THE INVENTION

**[0033]** Below, the present invention will be described by way of Examples.

**[0034]** The constituent components (A) to (D) and the relevant components employed Examples and Comparative Examples are summarized in Tables 1 to 5.

Table 1

Constituent Component (A)			
	A-1	A-2	a-1
For the entire (A)			
MFR (g/10 min.)	50	65	50
Ethylene content (wt. %)	5.2	5.1	5.1
Av. MFR of Fp (g/10 min.)	152	260	150
MFR of Fh (g/10 min.)	500	720	-
Content of Fh (wt. %)	35	35	-
Pentad fraction of Fp (%)	98.6	98.4	98.5
Intr. viscosity of Fc (dl/g)	8.1	8.8	8.0

Table 2

Constituent Component (A)			
	a-2	a-3	a-4
For the entire (A)			
MFR (g/10 min.)	52	64	51
Ethylene content (wt. %)	5.3	5.2	5.2
Av. MFR of Fp (g/10 min.)	147	270	155
MFR of Fh (g/10 min.)	300	1200	500
Cont. of Fh (wt. %)	35	35	35
Pentad fraction of Fp (%)	98.6	98.1	96.4
Intr. viscosity of Fc (dl/g)	8.2	8.3	7.9
Notes for Tables 1 and 2: MFR: Melt flow rate according to ASTM D 1238, determined at 230 °C under a load of 2160 g. Fp: Propylene homopolymer portion. Fh: Low molecular weight propylene homopolymer. Fc: Ethylene/propylene random copolymer portion. Cont. of Fh: $= (Fh/Fp) \times 100$ Pentad fraction of Fp: Determined as proportion of mmmm peaks to the entire absorption peaks within the methyl-carbon region in $^{13}\text{C}$ -NMR spectrum. Intr. viscosity: Intrinsic viscosity determined at 135 °C in decalin.			

Table 3

Constituent Component (B)					
	B-1	b-1	b-2	b-3	b-4
St. cont. (wt. %)	15	30	15	15	14
MFR (g/10 min.)	6	12	0.2	73	9
Hydr. degree (mole %)	99	99	99	98	0
Type of polymer	SEBS	SEBS	SEBS	SEBS	SIS
Noted for Table 3: St: Styrene SEBS: Styrene/ethylene-butene/styrene triblock-copolymer SIS: Styrene/isoprene/styrene triblock-copolymer					

Table 4

Constituent Component (C)						
	C-1	C-2	c-1	c-2	c-3	c-4
St. cont. (wt. %)	65	45	18	64	65	70
MFR (g/10 min.)	0.9	3.2	4.8	22	0.06	1.8
Hydr. deg. (mole %)	99	99	99	99	98	0
Type of polymer	SEPS	SEBS	SEBS	SEBS	SEBS	SBS
Noted for Table 3: St: Styrene SEPS: Styrene/ethylene-propylene/styrene triblock-copolymer SEBS: Styrene/ethylene-butene/styrene triblock-copolymer SBS: Styrene/butadiene/styrene triblock-copolymer						

Table 5

Constituent Component (D)			
	D-1	d-1	d-2
Average P.S. ( $\mu\text{m}$ )	2.4	7.5	1.2
Material	talc	talc	$\text{CaCO}_3$
Average P.S.: Average particle size was determined by a laser beam diffraction method.			

#### Examples 1 - 3 and Comparative Examples 1 - 14

**[0035]** Using the constituent components given in Tables 1 - 5, the crystalline propylene block-copolymer compositions given in Tables 6 - 8 were obtained. For these compositions, material properties were observed. The test specimens used for the testings were prepared using an injection molding machine of The Japan Steel Works, Ltd., Model J100SA 11, by injection-molding the composition at a cylinder set temperature of 230 °C and at a metal mold temperature of 40 °C. The results are shown in Tables 6 - 8.

Table 6

	Example			Compar. Example		
	1	2	3	1	2	3
Component (A) proportion (wt. %)	A-1 62	A-2 62	A-1 62	a-1 62	a-2 62	a-3 62
Component (B)	B-1	B-1	B-1	B-1	B-1	B-1

Table 6 (continued)

	Example			Compar. Example		
	1	2	3	1	2	3
proportion (wt. %)	14	14	10	14	14	14
Component (C)	C-1	C-1	C-2	C-1	C-1	C-1
proportion (wt. %)	4	4	8	4	4	4
Component (D)	D-1	D-1	D-1	D-1	D-1	D-1
proportion (wt. %)	20	20	20	20	20	20
FM (MPa)	2430	2410	2370	2440	2410	2350
IZ (J/m)	380	350	410	390	370	170
HR	74	73	73	74	73	72
BT (°C)	-32	-33	-30	-31	-32	-25
SFL (cm)	142	155	145	130	133	163

Table 7

	Comparative Example					
	4	5	6	7	8	9
Component (A)	a-4	A-1	A-1	A-1	A-1	A-1
proportion (wt. %)	62	62	62	62	62	62
Component (B)	B-1	b-1	b-2	b-3	b-4	B-1
proportion (wt. %)	14	14	14	14	14	14
Component (C)	C-1	C-1	C-1	C-1	C-1	c-1
proportion (wt. %)	4	4	4	4	4	4
Component (D)	D-1	D-1	D-1	D-1	D-1	D-1
proportion (wt. %)	20	20	20	20	20	20
FM (MPa)	2180	2590	2280	2430	2110	2280
IZ (J/m)	390	220	150	130	320	420
HR	65	76	69	74	62	68
BT (°C)	-31	-18	-25	-15	-20	-23
SFL (cm)	143	143	136	150	142	145

Table 8

	Comparative Example					
	10	11	12	13	14	15
Component (A)	A-1	A-1	A-1	A-1	A-1	A-1
proportion (wt. %)	62	62	62	62	62	62
Component (B)	B-1	B-1	B-1	B-1	B-1	B-1
proportion (wt. %)	14	14	14	14	14	18
Component (C)	c-2	c-3	c-4	C-1	C-1	-
proportion (wt. %)	4	4	4	4	4	-

Table 8 (continued)

	Comparative Example					
	10	11	12	13	14	15
Component (D) proportion (wt. %)	D-1 20	D-1 20	D-1 20	d-1 20	d-2 20	D-1 20
FM (MPa)	2390	2460	2480	2170	1980	2230
IZ (J/m)	100	160	210	250	380	390
HR	69	73	74	68	62	70
BT (°C)	-11	-22	-21	-25	-19	-23
SFL (cm)	132	151	142	139	146	146
Notes for Tables 6 to 8: FM: Flexural modulus, determined according to ASTM D 790. IZ: Izod impact strength, determined according to ASTM D 256 at -30 °C. HR: Surface hardness, determined according to ASTM D 685 using steel ball R, the values being represented in the R-scale. BT: Brittle temperature, determined according to ASTM D 746. SFL: Flowability, expressed by spiral flow length determined by carrying out an injection molding of the composition using an injection mold having a spiral flow path of a thickness of 3 mm, a width of 10 mm and a length of 2,000 mm and observing the flow length of the resin.						

**[0036]** From the results of Tables 6 to 8, it is seen that all the compositions of Examples 1 to 3 are superior in the flowability (SFL) and also in the balance between the flexural modulus (FM), Izod impact strength (IZ), surface hardness (HR) and brittle temperature (BT) as well.

**[0037]** On the contrary, the compositions of Comparative Examples 1 and 2 exhibit inferior flowabilities and the composition of Comparative Example 3 is inferior in the Izod impact strength and brittle temperature.

**[0038]** The composition of Comparative Example 4 is inferior in the flexural modulus and surface hardness. The composition of Comparative Example 5 is inferior in the Izod impact strength and brittle temperature. The composition of Comparative Example 6 is inferior in the Izod impact strength, surface hardness and flowability. The composition of Comparative Example 7 is inferior in the Izod impact strength and brittle temperature, the composition of Comparative Example 8 is inferior in the flexural modulus, surface hardness and brittle temperature and the composition of Comparative Example 9 is inferior in the surface hardness and brittle temperature.

**[0039]** The composition of Comparative Example 10 exhibits inferior values for the Izod impact strength, for the surface hardness, for the brittle temperature and for the flowability. The compositions of Comparative Examples 11 and 12 are inferior in the Izod impact strength and in the brittle temperature.

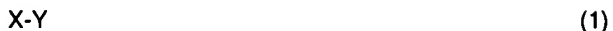
**[0040]** The composition of Comparative Example 13 is inferior in all the material properties. Comparative Examples 14 gave inferior results as to the flexural modulus, surface hardness and brittle temperature and Comparative Example 15 gave inferior results as to the flexural modulus and brittle temperature.

## Claims

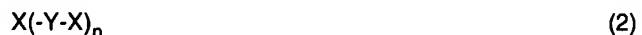
1. A crystalline propylene block-copolymer composition for use in automobile parts, comprising

- (A) 41 to 71% by weight of a crystalline propylene block-copolymer composed of a propylene homopolymer portion (Fp) and an ethylene/propylene random copolymer portion (Fc) and having an ethylene content of 1 to 10% by weight and a melt flow rate (MFR, determined at 230°C under a load of 2160 g) of 20 to 100 g/10 min.,
- (B) 12 to 22% by weight of a first hydrogenated product of a block-copolymer composed of blocks represented by the following formula (1) or (2), namely,





in which X stands for a polymer block of a vinyl-mono-substituted aromatic hydrocarbon, Y stands for an elastomeric polymer block of a conjugated diene and n is an integer of 1 to 5, wherein the content of X amounts to 5 to 22% by weight and the melt flow rate (MFR) for the entire block-copolymer is 0.5 to 70 g/10 min. and wherein the hydrogenation degree is at least 90 mole %, (C) 2 to 12% by weight of a second hydrogenated product of a block-copolymer composed of blocks represented by the following formula (1) or (2), namely,



in which X stands for a polymer block of a vinyl-mono-substituted aromatic hydrocarbon, Y stands for an elastomeric polymer block of a conjugated diene and n is an integer of 1 to 5, wherein the content of X amounts to 30 to 80% by weight and the melt flow rate (MFR) for the entire block-copolymer is 0.1 to 20 g/10 min. and wherein the hydrogenation degree is at least 90 mole %, and (D) 15 to 25% by weight of talc of a grade having an average particle size, determined by a laser beam diffraction method, of 1 to 5  $\mu\text{m}$ ,

wherein, in the crystalline propylene block-copolymer (A), the said propylene homopolymer portion (Fp) contains 20 to 50% by weight of a low molecular weight propylene homopolymer (Fh) having a melt flow rate (MFR, determined at 230°C under a load of 2160 g) of 400 to 1,000 g/10 min. and is constituted of a propylene homopolymer having an average melt flow rate (MFR, determined at 230°C under a load of 2160 g), for the entire propylene homopolymer portion (Fp), of 100 to 350 g/10 min. and an isotactic pentad fraction (mmmm), determined by  $^{13}\text{C}$ -NMR, of at least 98% and the said ethylene/propylene random copolymer (Fc) is constituted of an ethylene/propylene random copolymer having an intrinsic viscosity  $[\eta]$ , determined at 135°C in decalin, of 7 dl/g or higher.

2. A crystalline propylene block-copolymer composition according to claim 1, wherein the vinyl-mono-substituted aromatic hydrocarbon constituting the polymer block represented by X of the formula (1) or (2) in the first and second hydrogenation products of block-copolymers (B) and (C) is constituted of one or more members selected from styrene,  $\alpha$ -methylstyrene, p-methylstyrene, chlorostyrene, lower alkyl-substituted styrenes and vinyl-naphthalene.
3. A crystalline propylene block-copolymer composition according to claim 1 or claim 2, wherein the conjugated diene constituting the elastomeric polymer block represented by Y of the formula (1) or (2) in the first and second hydrogenation products of block-copolymers (B) and (C) is constituted of one or more members selected from butadiene, isoprene and chloroprene.
4. A crystalline propylene block-copolymer composition according to any one of the preceding claims, wherein the first and second hydrogenation products of block-copolymers (B) and (C) are each constituted of one or more members selected from styrene/ethylene-butene/styrene triblock-copolymers (SEBS), styrene/ethylene-propylene/styrene triblock-copolymers (SEPS) and styrene/ethylene-propylene diblock copolymers (SEP).
5. Use of a crystalline propylene block-copolymer composition as claimed in any one of claims 1 to 4 as an injection molding material.
6. An automobile part formed from a crystalline block-copolymer composition as claimed in any one of claims 1 to 4.

## Patentansprüche

1. Zusammensetzung mit kristallinem Propylen-Blockcopolymer zur Verwendung bei Kraftfahrzeugteilen, aufweisend

A) 41 bis 71 Gewichts% eines kristallinen Propylen-Blockcopolymer, das aus einem Propylen-Homopolymer-Segment (Fp) und einem statistischem Copolymer-Segment aus Ethylen-Propylen (Fc) besteht und einen Ethylen-Gehalt von 1 bis 10 Gewichts% und eine Schmelzflussrate (MFR (melt flow rate), bestimmt bei 230° C unter einer Belastung von 2160 g) von 20 bis 100 g/10 min hat,

B) 12 bis 22 Gewichts% eines ersten hydrierten Produkts eines Blockcopolymer, das aus Blöcken besteht, die durch die folgende Formel (1) oder (2) dargestellt werden, nämlich



worin X für einen Polymerblock aus einem Vinyl-monosubstituierten aromatischen Kohlenwasserstoff steht, Y für einen elastomeren Polymerblock aus einem konjugierten Dien steht und n eine ganze Zahl von 1 - 5 ist, wobei der Gehalt von X 5 bis 22 Gewichts% beträgt und die Schmelzflussrate (MFR) für das gesamte Blockcopolymer 0,5 - 70 g/10 min ist, und wobei der Hydrierungsgrad mindestens 90 Mol% ist,

C) 2 - 12 Gewichts% eines zweiten hydrierten Produkts eines Blockcopolymer, das aus Blöcken besteht, die durch die folgende Formel (1) oder (2) dargestellt werden, nämlich



worin X für einen Polymerblock aus einem Vinyl-monosubstituierten aromatischen Kohlenwasserstoff steht, Y für einen elastomeren Polymerblock aus einem konjugierten Dien steht, und n eine ganze Zahl von 1 - 5 ist, wobei der Gehalt von X 30 bis 80 Gewichts% beträgt und die Schmelzflussrate (MER) für das gesamte Blockcopolymer 0,1 - 20 g/10 min ist, und wobei der Hydrierungsgrad mindestens 90 Mol% ist, und

D) 15 bis 25 Gewichts% Talk einer Körnung mit einer mittleren Teilchengröße, bestimmt durch Laserstrahl-Diffraktionsverfahren, von 1 - 5 µm,

wobei in dem kristallinen Propylen-Blockcopolymer (A) das Propylen-Homopolymer-Segment (Fp) 20 bis 50 Gewichts% eines Propylen-Homopolymers (Fh) einer geringen Molekülmasse mit einer Schmelzflussrate (MFR, bestimmt bei 230° C unter einer Belastung von 2160 g) von 400 - 1000 g/10 min enthält, und aus einem Propylen-Homopolymer mit einer mittleren Schmelzflussrate (MFR, bestimmt bei 230° C unter einer Belastung von 2160 g) für das gesamte Propylen-Homopolymer-Segment (Fp) von 100 - 350 g/10 min, und einem isotaktischen Pentad-Anteil (mmmm), bestimmt durch <sup>13</sup>C-NMR, von mindestens 98% gebildet ist, und das statistische Ethylen/Propylen-Copolymer (Fc) aus einem statistischen Ethylen/Propylen-Copolymer mit einer inneren Viskosität [η], bestimmt bei 135° C in Decalin, von 7 dl/g oder höher gebildet ist.

2. Zusammensetzung mit kristallinem Propylen-Blockcopolymer nach Anspruch 1, worin der Vinyl-monosubstituierte aromatische Kohlenwasserstoff, der den Polymerblock bildet, der durch X der Formel (1) oder (2) in dem ersten Hydrierungsprodukt und dem zweiten Hydrierungsprodukt der Blockcopolymere (B) und (C) dargestellt wird, aus einem oder mehr Gliedern gebildet wird, die ausgewählt sind aus Styrol, α-Methylstyrol, p-Methylstyrol, Chlorstyrol, mit niedrigeren Alkylen substituierten Styrolen und Vinylnaphthalin.

3. Zusammensetzung mit kristallinem Propylen-Blockcopolymer nach Anspruch 1 oder Anspruch 2, worin das konjugierte Dien, das den elastomeren Polymerblock bildet, der durch Y der Formel (1) oder (2) in dem ersten Hy-

drierungsprodukt und dem zweiten Hydrierungsprodukt der Blockcopolymere (B) und (C) dargestellt wird, von einem oder mehreren Gliedern gebildet wird, die ausgewählt sind aus Butadien, Isopren und Chloropren.

4. Zusammensetzung mit kristallinem Propylen-Blockcopolymer nach einem der vorangehenden Ansprüche, worin das erste und zweite Hydrierungsprodukt der Blockcopolymere (B) und (C) jeweils von einem oder mehreren Gliedern gebildet werden, die ausgewählt sind aus Styrol/Ethylen-Buten/Styrol-Triblockcopolymeren (SEBS), Styrol/Ethylen-Propylen/Styrol-Triblockcopolymeren (SEPS) und Styrol/Ethylen-Propylen-Diblockcopolymeren (SEP).
5. Verwendung einer Zusammensetzung mit kristallinem Propylen-Blockcopolymer, wie sie in einem der Ansprüche 1 bis 4 beansprucht ist, als ein Spritzgießmaterial.
6. Kraftfahrzeugteil, hergestellt aus einer Zusammensetzung mit kristallinem Blockcopolymer wie sie in einem der Ansprüche 1 bis 4 beansprucht ist.

## Revendications

1. Composition de copolymère séquencé de propylène cristallin à utiliser dans des pièces automobiles, comprenant :

(A) 41 à 71 % en poids d'un copolymère séquencé de propylène cristallin composé d'une portion de type homopolymère de propylène (Fp) et d'une portion de copolymère aléatoire éthylène/propylène (Fc) et ayant une teneur en éthylène de 1 à 10 % en poids et une vitesse d'écoulement de la matière fondue (MFR, déterminée à 230°C sous une charge de 2160 g) de 20 à 100 g/10 min,

(B) 12 à 22 % en poids d'un premier produit hydrogéné d'un copolymère séquencé composé de séquences représentées par la formule (1) ou (2) suivante, c'est à dire,



dans laquelle X représente une séquence polymère d'un hydrocarbure aromatique monosubstitué par un vinyle, Y représente une séquence de polymère élastomérique d'un diène conjugué et n est un entier de 1 à 5, dans laquelle la concentration de X s'élève à 5 à 22 % en poids et la vitesse d'écoulement de matière fondue (MFR) pour l'ensemble du copolymère séquencé est de 0,5 à 70 g/10 min et dans laquelle le degré d'hydrogénation est d'au moins 90 % en mole,

(C) 2 à 12 % en poids d'un second produit hydrogéné d'un copolymère séquencé composé de séquences représentées par la formule (1) ou (2) suivante, c'est à dire,



dans laquelle X représente une séquence polymère d'un hydrocarbure aromatique monosubstitué par un vinyle, Y représente une séquence de polymère élastomérique d'un diène conjugué et n est un entier de 1 à 5, dans laquelle la concentration de X s'élève à 30 à 80 % en poids et la vitesse d'écoulement de matière fondue (MFR) pour l'ensemble du copolymère séquencé est de 0,1 à 20 g/10 min et dans laquelle le degré d'hydrogénation est d'au moins 90 % en mole, et

(D) 15 à 25 % en poids de talc d'une qualité ayant une taille moyenne de particule, déterminée par une méthode de diffraction d'un faisceau laser, de 1 à 5 µm,

dans laquelle, dans le copolymère séquencé de propylène cristallin (A), ladite portion d'homopolymère (Fp) contient 20 à 50 % en poids d'un homopolymère de propylène de bas poids moléculaire (Fh) ayant une vitesse d'écou-

lement de matière fondue (MFR, déterminée à 230°C sous une charge de 2160 g) de 400 à 1 000 g/10 min, et est constituée d'un homopolymère de propylène ayant une vitesse moyenne d'écoulement de matière fondue (MFR, déterminée à 230°C sous une charge de 2160 g), pour l'ensemble de la portion homopolymère de propylène (Fp), de 100 à 350 g/10 min et une fraction isotactique d'ordre cinq (mmmm), déterminée par <sup>13</sup>C-RMN, d'au moins 98 % et ledit copolymère aléatoire éthylène/propylène (Fc) est constitué d'un copolymère aléatoire éthylène/propylène ayant une viscosité intrinsèque [η], déterminée à 135°C dans la décaline, de 7 dl/g ou supérieure.

2. Composition de copolymère séquencé de propylène cristallin selon la revendication 1, dans laquelle l'hydrocarbure aromatique monosubstitué par un vinyle constituant la séquence polymère représentée par X de la formule (1) ou (2) dans le premier et second produits d'hydrogénation de copolymère séquencés (B) et (C) est constitué d'un ou plusieurs éléments choisis parmi le styrène, l' $\alpha$ -méthylstyrène, le p-méthylstyrène, le chlorostyrène, les styrènes substitués par un alkyle inférieur et le vinyl-naphtalène.
3. Composition de copolymère séquencé de propylène cristallin selon la revendication 1 ou la revendication 2, dans laquelle le diène conjugué constituant la séquence polymère élastomérique représentée par Y de la formule (1) ou (2) dans le premier et second produits d'hydrogénation de copolymères séquencés (B) et (C) est constitué d'un ou plusieurs éléments choisis parmi le butadiène, l'isoprène et le chloroprène.
4. Composition de copolymère séquencé de propylène cristallin selon l'une quelconque des revendications précédentes, dans laquelle le premier et second produits d'hydrogénation de copolymères séquencés (B) et (C) sont chacun constitués d'un ou plusieurs éléments choisis parmi les copolymères triséquencés styrène/éthylène-butène/styrène (SEBS), les copolymères triséquencés styrène/éthylène-propylène/styrène (SEPS) et les copolymères diséquencés styrène/éthylène-propylène (SEP).
5. Utilisation d'une composition copolymère de séquencé de propylène cristallin telle que revendiquée dans l'une quelconque des revendications 1 à 4 comme matériau de moulage par injection.
6. Pièce automobile formée à partir d'une composition de copolymère séquencé de propylène cristallin telle que revendiquée dans l'une quelconque des revendications 1 à 4.